Positron Annihilation Lifetime Study on free Volume Changes in TLD by Gamma – irradiation

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Abstract

Positron annihilation lifetime has been utilized for the first time to investigate the free – volume hole properties in thermolumenscent dosimeter (TLD) as a function of gamma-dose . The hole volume, free volume fraction determined form orthopsitronium lifetime are found to be dramatically increase to large values , and then to minimum values as a function of gamma-dose . The free – volume holes size is found to be $0.163nm^3$ and to have maximum of $0.166nm^3$ at the gamma-dose of 0.1 and 0.8 Gy, respectively.

Introduction

Studies of the microstructure free- volume properties at a molecular and atomic scale can provide a basic understanding of the mechanical and physical properties of polymers(1). In PAL technique [2], one employs the positron emitted from a radioactive source as a probe to monitor the lifetime of the positron and positronium (Ps) (a bound atom consists of an electron and a positron) in polymer material under study. Because of the possitive –charged nature of the positron, the positron and the positronium atom are attracted by the core of electrons of the polymers and trapped in open spaces like holes voids, or cavities. The annihilation photons come mainly from these open spaces . Positrons are commonaly produced by positron radioactive isotope , such as ^{22}Na. The Ps atom formed can be in the singlet (P-Ps) or triplet state (o-Ps) , with mean lifetime

in free spaces of 0.125 ns and 140 ns , respectively. In condensed medium the mean lifetime of o-Ps is considerably reduced to a few nanosecond [3] due to its interaction with surrounding molecules of the medium. The results for o-Ps lifetime and its probability are related to free-volume hole size (V_h) , fractional free volume (F_h) and distribution. The relation between the mean radius of free –volume hole size and the o-P_s lifetime τ_{o-Ps} was found according to the semi -emprical formula (4):

$$\tau_{0-P_s} = 0.5 \left[\frac{1 - R/R_O +}{0.159 \sin\left(\frac{2\pi R}{R_O}\right)} \right]^{-1} ns \ldots\ldots\ldots(1)$$

where $R = R_o + \Delta R, \Delta R$ is the electron layer thickness The free-volume –hole size (V_h) is :

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$$V_h = \frac{4}{3}\pi R^3 \dots\dots\dots\dots(2)$$

The fractional free volume (the ratio of the free volume to the total volume) was found according to fitted empirical formula [5]as:

$$F_h = AV_h I_{0-P_s} \dots\dots\dots\dots\dots(3)$$

where A is a constant , its value is between 1-2 for polymer , V_h is the free-volume hole size and I_{0-P_s} is the intensity of τ_{0-P_s} . PAL is widely used for investigation different aspects of polymer properties [6-11]. Positron and positronium lifetime in polymer material may affected by different factors such as changing in degree of crystallinity, blending of polymer, Plasticization of polymer aging of polymer , temperature , irradiation of polymer [12-20]. When an organic materials such as polymer is irradiated by neutron-flux , different processes can take place in the materials according to the energy of the neutron . Within the reaction with the nuclei of the atoms of the irradiated medium. Fast neutrons transfer a considerable a mount of their energy. Each struck nucleus is ejected as a recoil ion that interact with orbital electrons of the medium, thus production molecular excitation and ionization. The initial neutrons are degraded to a thermal – energy state as a result of the several scatterings. Irradiation effects on PTFE were studied by kindl et al [16] using gamma –irradiation in the dose range up to 200 kGy. They found that τ_{0-P_s} life to be decreased with increasing $\gamma - dose$ as a result of increasing crystallinity and radicals quenching .Kindl[17] found that free radicals on the one hand and irradiation induces structure alterations on the other hand change the positron lifetime parameters within the irradiation dose range up to 200 kGy. The studies of Consolati and Quasso [19] showed that

the longest lifetime component is certainly due to o-Ps "Pick off" annihilation. Recently Al-Bayati [20] found that γ-irradiation induces structural changes in PTFE, increasing crystallinity in the amorphous regions within the irradiation dose up to 887 kGy. Despite of numerous studies on positron annihilation lifetime in polymers to the best of our knowledge no one investigate the effect of absorbed dose on TLD structure ,hence on its efficiency for absorbed dose measurement. Therefore the present work is the first attempt to study the effect of γ-dose on V_f and F_r of TLD ,hence TLD structure.

Experimental Procedure and Data Analysis

The position lifetime measurement were conducted using fast slow coincidence system , have a time resolution of 340 ps . The position source consisted of 12 µCi of ^{22}NaCl placed between two aluminum foils of 5 µm thickness the fraction of positrons absorbed in the source was found to be 8% using the recent formula of Al-bayati [12] : $\alpha_+ / \rho = 27.84 Z^{0.137}$ .The TLD samples used for present study , are the same as those used as irradiation dosimetry , (it contains 70% PTFE and 30% LiF) with diameter of 1.3 cm and thickness of 0.1 mm supplied by Vinten . The irradiation was done with CO-60 in air at R.T. , with a dose rate of 0.1 Gy/min up to a total dose of 2Gy . The lifetime spectra were measured for each individual dose value with a total integral counts of 2×10^6 . The peak – to – background ratio was better than 2300:1. The lifetime spectra were analyzed in three-lifetime components using PFPOSFIT program [21] include two components for source correction .

The lifetime components , their relative intensities and the parameters of the prompt curve , were simultaneously fitted .

Results and Discussion

The average radius of the free – volume hole , the free volume hole size and the free volume fraction , were calculated using equations (1) and (2) , respectively. The o-ps lifetime (τ_3) and the second lifetime (τ_2), free volume V_f and free volume fraction (F_r) were plotted as a function of γ - dose in Figs.(1) , (2) and (3) , respectively . It is clear that (τ_2), which is related to free positron annihilation , and which its decay depends less on the structural state of the polymer , was found to be γ - dose independent , because no influence of γ - dose is expected on the mean electron density . The longest lifetime component τ_3, which is due to o-p$_s$ "pick off" annihilation was found to be 2578 ps for unirradated samples , corresponding to V_f of 0.154 nm^3 and F_r of 0.94% . The behaviour of V_f hence τ_3 and F_r with γ - dose can be divided into two stages as shown in Figs.(1) , (2) and (3) . In the first stage up to 0.8 Gy , the V_f hence τ_3 , and F_r increase with increasing dose and reached an increment of 6.1% , 3.4% and 2.7% , respectively at 0.1 Gy , then reached a maximum increment of 8.0% , 4.4% and 20% , respectively for V_f , τ_3 and F_r at 0.8 Gy . In the second stage between (0.8 and 2) Gy , V_f hence τ_3 and F_r begin to decrease gradually with increasing dose and reached a minimum increment of 1.41 , 0.7% and 1.9% respectively . These results suggest that γ - induces structural changes in TLD resulting in degradation of TLD chains during the first irradiation stage , which causes an increase of the three parameters . As

the dose increases above 0.8Gy , the effective free radicals induced by γ - irradiation , and increasing crystallinity in the amorphous regions of TLD samples , resulting in reduction of free volume fraction , free volume hole size , hence o-p$_s$ lifetime .
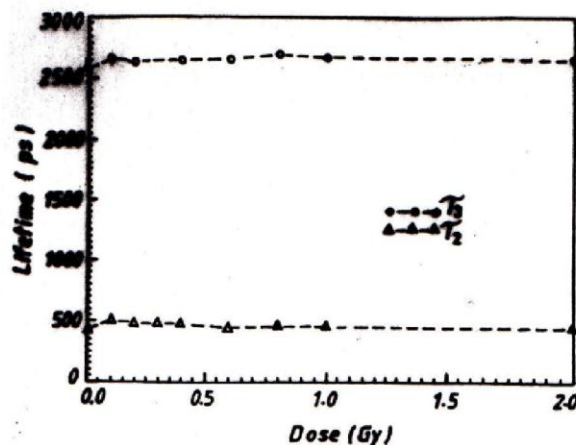

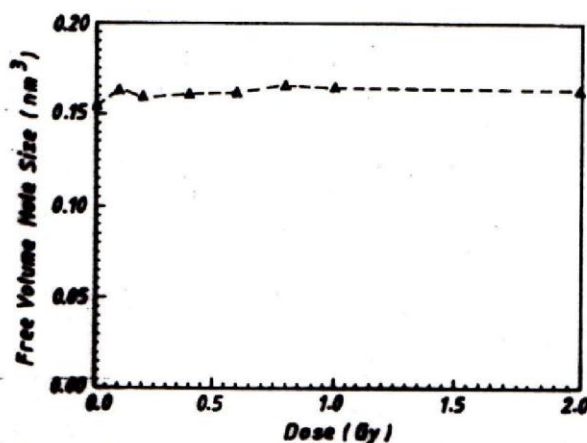Fig. (1) Gamma – ray irradiation effects on T_3 & T_2

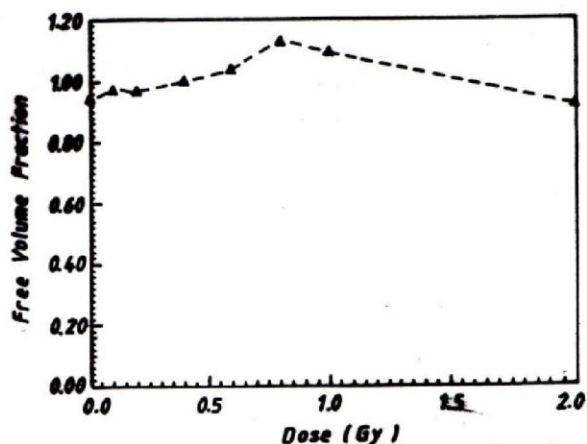Fig. (2) Gamma–ray irradiation effects on free volume hole size in TLD

Fig. (3) Gamma - ray irradiation effects on free volume fraction in TLD

Conclusion

Degradation of TLD chains caused by gamma – irradiation results in an increment of 10% and 25% per Gy in V_f and F_r , respectively , which may reduce significantly the efficiency of TLD for the received dose measurement for each usage .

Acknowledgment

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دراسة زمن عمر فناء البوزترون لتغيرات الحجم الحر
في مقياس الوميض الحراري الضوئي المشعع بكاما

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خلاصة

تم استخدام زمن عمر فناء البوزترون لاول مرة لدراسة خصائص الحجم الحر فـي مقيـاس الوميـض
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القيمة ١٦٣, ٠ نانومتر مكعب له قيمة عظمى هي ١٦٦, ٠ نانومتر مكعب عند جرع كامــا ١, ٠ و ٨, ٠
كراي على التوالي .